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Recent trends in ozone in the upper stratosphere: Implications for chlorine chemistry

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Abstract. We have studied the implications of recent trends in the annual mean and the amplitude of the annual harmonic of ozone in the upper stratosphere from the 15 years of the combined data from the Nimbus-7 SBUV and the NOAA-11 SBUV/2 instruments. This was done in the context of the GSFC 2D model predictions of these trends which are based on plausible scenarios of anthropogenic Cly increase in the atmosphere. The comparison of the observed and model-estimated annual mean ozone trends show some similarity in their latitude and altitude characteristics. Both the model and data show a maximum ozone decrease of -6 to -10 % per decade at high latitudes in the upper stratosphere. However, there are also significant differences between the observed and computed trends which may be related to both the model uncertainty and the uncertainty in correcting for the long term instrument drift. The observations also suggest a decrease of 10-25 % per decade in the annual amplitude of ozone at 2 mb between 40°-60° in both hemispheres, with a relatively larger interannual variability in the northern hemisphere. These values are in general agreement with the model predictions and thus provide additional support in favor of the chlorine induced changes in ozone in the upper stratosphere.

Introduction

Recently, Hood et al. (1993) have reported a decrease of 6-12 % in upper stratospheric ozone at middle and high latitudes over an eleven year period from 1979 to 1990. Their study, based on the version 6 SBUV data from the Nimbus-7 satellite, supports an earlier conclusion by Hilsenrath et al. (1993) that ozone trends in the upper stratosphere are statistically significant and may have been caused by an increase in the chlorine level of the atmosphere related to anthropogenic perturbations. DeLuisi et al. (1994) have shown that the trends derived from the SBUV data over this period are in general agreement with those derived from the ozone profiles from the ground based Umkher measurements. The SBUV trends also show general similarity with the trends derived from the 2D photochemical models (WMO, 1991). These trends, however, are significantly larger than the trends reported by McCormick et al. (1992) from their analysis of SAGE I and SAGE II ozone profiles for the period February 1979 to April 1991.

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More recently, Chandra and McPeters (1994) have extended the ozone data base by combining the Nimbus-7 SBUV data with the NOAA-11 SBUV/2 data from January 1989 to December 1993. Though the combined data set covering a 15 year period does not significantly alter the length of the time series for studying long term changes in stratospheric ozone, it is the best data set available to study the various implications of long term changes in the stratosphere. It has been shown by Chandra et al.(1993, to be referred to as C93) that both the annual mean and the amplitude of the annual harmonic of ozone are influenced by the chlorine catalytic loss cycle. The annual harmonic at middle and high latitudes accounts for more than 80 % of the total variance of the annual wave (see also Perliski and London, 1989). A study of long term change in the annual cycle of ozone thus provides an additional constraint in delineating the role of odd chlorine in the stratosphere. The purpose of this paper is to study long term changes in the annual mean and the annual amplitude of ozone in the upper stratosphere at middle and high latitudes in the context of the GSFC 2D model results as discussed in C93.

Data Description

The data used in this study are based on the monthly values of ozone time series from the Nimbus-7 SBUV and the NOAA-11 SBUV/2 instruments from January 1979 to December 1993 as discussed in Chandra and McPeters (1994). They respectively cover periods from January 1979 to December 1989 and January 1989 to December 1993 with an overlapping period of one year. To account for the differences in the absolute calibration of the two instruments, the SBUV time series have been normalized in terms of the SBUV/2. The normalization is achieved by readjusting the SBUV time series by a factor calculated from the mean of the ratio of the SBUV and SBUV/2 values over the overlapping period. The normalization factor varies from about 4-6 % at and above 2 mb to about -1 to -5% at 10 mb and below. The time series for each year are Fourier analyzed to calculate the annual mean and the annual, semiannual, and terannual amplitudes and phases. Our discussions will be limited to the annual mean and annual wave at middle and high latitudes where changes in levels of atmospheric chlorine have their largest effects.

We have also performed a Fourier analysis on the analogous NMC temperature time series. The annual cycles of ozone in the upper stratosphere are driven by the annual cycles of temperature through temperature dependent loss rates. The interannual variability and long term changes in ozone are, therefore, expected to be closely coupled with the interannual variability and long term changes in temperature. Unfortunately, the long term changes in the NMC temperature in the upper stratosphere may have an uncertainty of 1-3 K because of the several adjustments made in the temperature time series using rocket measurements (Finger et al., 1993). Such adjustments may also have affected the annual amplitude of temperature since some of the adjustments were made during the middle of the year.

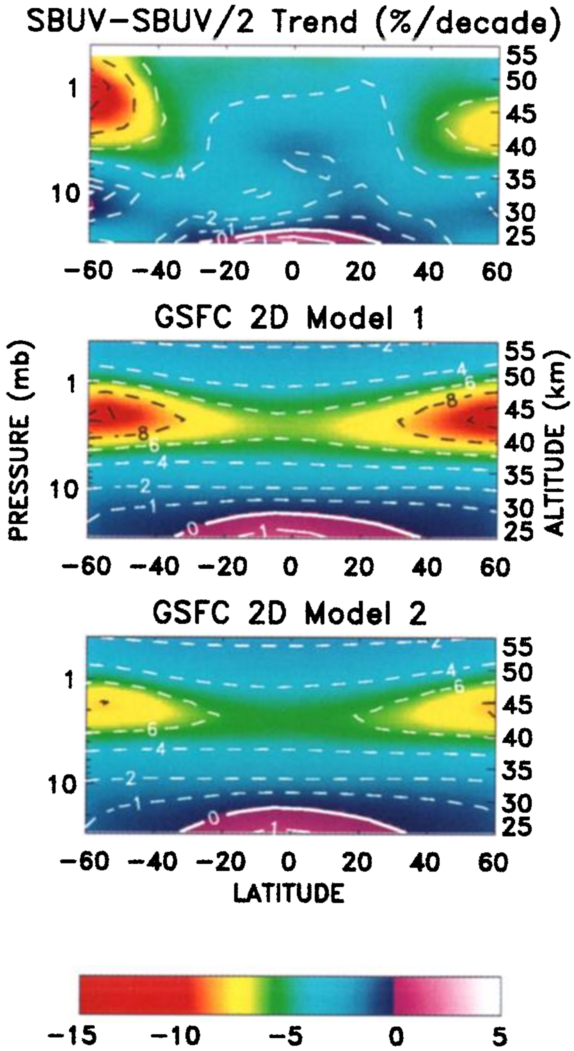

Figure 1. Comparison of the decadel trend in ozone (% per decade) inferred from the SBUV and the SBUV/2 data (upper panel) and trends estimated from the GSFC 2D model (middle and lower panel) as discussed in the text.

2D Model Implications of the Annual Mean Ozone Trend

Figure 1 compares the observed trend in ozone from the 1979 to 1993 (expressed as % change per decade) from the combined SBUV and NOAA-11 data (upper panel) with the estimated trend for this period based on the GSFC 2D model simulations (middle and lower panels). The observed trend is derived from the linear regression analysis of the annual mean ozone time series. The model assumes the total chlorine in the source gases at the ground to be about 2.5 ppbv and 3.5 ppbv, respectively, for 1980 and 1990 conditions (WMO, 1991)which reflects a chlorine increase of about 1 ppbv/decade. The middle panel in Figure 1 uses 'standard chemistry' (DeMore et al., 1992), without modifying the partitioning of Cly (model 1), and the lower panel assumes a Cly partitioning which includes the reaction $ClO+OH \rightarrow HCl+O_2$ with a branching ratio of 0.07 discussed in C93 (model 2). The reaction $ClO+OH \rightarrow HCl+O_2$ (McElroy and Salawitch, 1989) in model simulations changes the Cly family partitioning by increasing reservoir chlorine (HCl) at the expense of active chlorine (ClO). As discussed in C93, this reduces the difference between the observed and calculated values of both the annual mean and the annual amplitude of ozone. For example, at mid and high latitudes at 2 mb, the model with 'standard chemistry' tends to underestimate the observed values of ozone by about 30 % and the observed values of annual amplitude by about a factor of 2-3. The inclusion of the $ClO+OH \rightarrow HCl+O_2$ reaction reduces the difference between the observed and calculated values ozone to about 15-20 %. A similar improvement is also seen between the calculated and the observed values of the annual amplitude of ozone. This change in partitioning also reduces the influence of Cly changes in producing long term changes in ozone in the upper stratosphere (Toumi and Bekki, 1993).

The basic pattern of the observed trends in Figure 1 is similar to the one derived from the weekly time series of Umkher layers using the same data set (S. Hollandsworth, personal communication, 1994) instead of the annual mean of ozone mixing ratio used in this study. In the upper stratosphere, which is the focus of this study, the calculated trend from the 2D model simulations in Figure 1 are symmetric with respect to the equator. The peak values are centered at about 2 mb in both models 1 and 2. In model 1 (middle panel), the trends vary from about -6 % at the equator to about -8 to -10 % per decade at high latitudes (50°-60°). In model 2, these values are reduced to about -5 % at low latitudes to about -6 to -8% per decade at high latitudes. The model results, when compared with the observed trends in Figure 1, show certain similarities with respect to their latitudinal characteristics. For example, the observed trend at 2 mb varies from about -3 to -5% per decade at low latitudes to about -6 to -10 % at high latitudes. The overall difference in the predicted trends with and without changing the Cly partitioning is relatively small. However, model 2 tends to support a smaller negative trend than that inferred from the data in the southern hemisphere.

There are several differences between the observed and model trends. The observed trends at low latitudes are statistically not significant and are smaller than indicated by the models. At mid-latitudes, the observed trends are not symmetrical with respect to the equator and vary more slowly with height than indicated by the models, particularly in the southern hemisphere. Some of the differences between the observed and calculated trends in ozone may be related to the model uncertainty, the poor vertical resolution of the SBUV and the SBUV/2 data, and the uncertainty in correcting for the instrument bias.

Trends in the Annual Mean and Annual Amplitude

The yearly changes in the annual mean and the annual amplitude at 2 mb, typical of the latitude region 40°-60°, are shown in Figure 2. Figure 2 compares the yearly changes in the annual mean and the annual amplitude of ozone at 2 mb both in the northern and the southern hemisphere, 50 °N (upper panel) and 50 °S (lower panel), from 1979 to 1993. The annual mean in both hemispheres shows a rapid

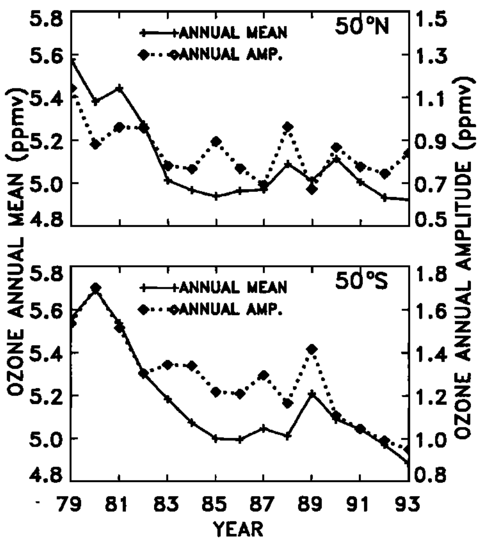

Figure 2. Time series of annual mean and annual amplitude of ozone mixing ratio at 2 mb showing their annual variability and long term trends at 50°N (upper panel) and 50°S(lower panel). The 93 data points may have some measurement uncertainty because the drift of the NOAA-11 orbits to the terminator.

decrease in ozone from 1979-80 to 1986 followed by a relatively slower decrease from 1986 to 1993. Since these periods correspond to the declining phase of the solar cycle 21 and the rising phase of solar cycle 22, the change in slope may be attributed to the solar cycle modulation of long term trends (negative)in ozone. A relatively larger slope during solar cycle 21 may also result from the deficiency in the version 6 algorithm in accounting for the long term instrument drift in the SBUV instrument. If the long term instrument drift is underestimated, it will tend to accentuate both the solar cycle component and the long term trend in ozone (Chandra and McPeters, 1994). Despite these uncertainties, the downward trend in ozone at 2 mb is generally consistent with the 2D model predictions based on Cly increases in the atmosphere. For example, the downward trends at 50°S and 50°N (Figure 2) in the annual mean of ozone inferred from a linear regression analysis are respectively -8.6$\pm$3.1 and -6.9$\pm$3.2 % per decade. These values are in generally good agreement with the 2D model predictions of 6-10 % decrease in ozone over this period (Figure 1) based on the plausible scenarios of Cly increase in the atmosphere (as discussed earlier in this paper).

Figure 2 also shows a downward trend in the annual amplitude of ozone at 2 mb in both the hemispheres modulated by interannual variability with periods of 2-3 years. The annual cycle in ozone in the upper stratosphere is primarily driven by a radiatively forced annual cycle in temperature through the temperature dependent loss rates of odd oxygen. Temperature, in addition to radiative forcing, is subject to interannual variability of dynamical origin (e.g, planetary wave forcing and stratospheric warmings). An increase (decrease) in the annual amplitude and the annual mean of temperature will cause a corresponding increase (decrease) in the annual amplitude of ozone and decrease (increase) in the annual mean of ozone. Their relative variability, of course, will depend upon the ozone sensitivity to temperature which is a complex function of various catalytic loss processes of odd oxygen involving odd hydrogen, nitrogen and chlorine(see, e.g., Stolarski and Douglass, 1985).

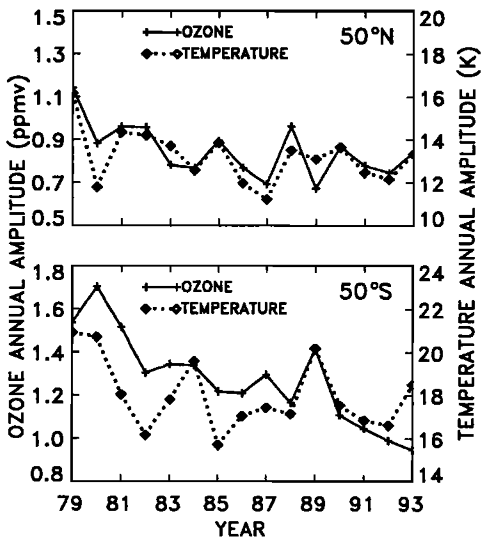

Figure 3a. The same as Figure 1 except the time series represent annual amplitudes of ozone and temperature.

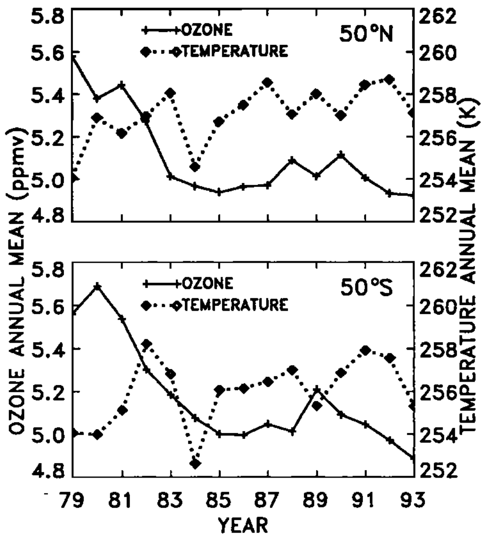

Figure 3b. The same as Figure 1 except the time series represent annual mean of ozone and temperature.

The relative changes in the annual amplitudes and the annual mean of ozone and temperature are illustrated in Figures 3a and 3b. Figure 3a shows a strong coupling between the annual amplitudes of ozone and temperature at 50°N and a relatively weaker coupling at 50°S over the 15 year period of the two time series. The correlation coefficients between the two time series are respectively 0.80 and 0.65. The correlation between the ozone and temperature time series are less apparent in the annual mean (Figure 3b) and may, in part, be related to the instrument bias in the SBUV instrument and the adjustments made in the NMC time series at various times (Finger et al., 1993) using rocket data. These figures indicate a negative trend in the annual amplitude and a positive trend in the annual mean of temperature in both hemispheres in the range of 1-2 K /decade which are not statistically significant. Based on this analysis, the negative trends in the annual mean and the annual amplitude of ozone discussed earlier in reference to Figure 2 do not appear to be caused by temperature.

The decadel trends in the annual amplitudes of ozone (Figure 2) are -26 $\pm$ 8.2 % and -16$\pm$13.7% at 50°S and 50°N respectively (all error bars are given at the 2 σ level)

A larger error bar at 50°N is due to a relatively larger interannual variability induced by planetary waves in the northern hemisphere. Such an error bar is typical of mid latitudes in the northern hemisphere and makes the inferred annual amplitude trend only marginally significant. In comparison, the annual amplitude in the southern hemisphere is statistically significant at latitudes between 40° and 60°S. These observations are in qualitative agreement with trends inferred from photochemical theory based on plausible scenarios of anthropogenic increases in Cly (WMO, 1991). For example, the decadel decrease in the annual amplitude of ozone computed from the 2D model at mid-latitudes is about 20-25 % for model 1 and 10-15 % for model 2. Since the 2D model uses a climatological temperature field, it cannot reproduce hemispherical asymmetries in the observed ozone trend induced by hemispherical asymmetries in the interannual variability of temperature. Even with these limitations, the evidence of Cly induced changes in upper stratospheric ozone is very compelling.

Summary and Conclusions

In this paper we have studied the implications of recent trends in ozone in the upper stratosphere using 15 years (1979-1993) of combined data from the Nimbus-7 SBUV and the NOAA-11 SBUV/2 instruments in the context of 2D model predictions of these trends. The model assumes two scenarios, (1) standard chemistry with no modification of the Cly partitioning, and (2) changing the Cly partitioning by including the reaction $ClO+OH \rightarrow HCl+O_2$ with a branching ratio of 0.07 discussed in C93. The comparison of the observed and model estimated trends in the annual mean of ozone show some similarity in the latitude and altitude characteristics. In the upper stratosphere both models and data show maximum decrease at high latitudes. The observed trends in the annual mean are in the range of -6 to -10 % per decade which are comparable to the estimated trends of both model 1 and model 2. Model 2 may still provide a better overall representation of ozone changes in the upper stratosphere because it tends to minimize the differences between the observed and calculated values of the annual mean and annual amplitude of ozone, as well as the ClO/HCl ratio as discussed in C93 and Toumi and Bekki (1993).

For the trends in the annual amplitude of ozone, both the model and observations suggest a decrease of 10-25% at 2 mb between 40°-60° in both hemispheres. The northern hemisphere observed trends are only marginally significant because of relatively larger interannual variability associated with the wave activity in this region. The southern hemisphere observations are statistically significant and qualitatively agree with photochemical theory, therefore providing additional evidence for the chlorine induced changes in upper stratospheric ozone.

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